Review Article

Research Progress on Durability of Cellulose Fiber-Reinforced Cement-Based Composites

Jie Liu¹ and Chun Lv^{ID}²

¹College of Light-Industry and Textile Engineering, Qiqihar University, Qiqihar 161006, China

²College of Architecture and Civil Engineering, Qiqihar University, Qiqihar 161006, China

Correspondence should be addressed to Chun Lv; lvchun8603@163.com

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The performance of cellulose fiber-reinforced cement-based composites (CFCCs) depends not only on the characteristics of the cement matrix and fibers but also on the bonding property of the matrix and fibers. The durability of cement-based composites including various properties such as impermeability, frost resistance, and carbonization resistance has an important impact on the long-term service life of the matrix structure. The presence of a large number of hydroxyl groups on the molecular chain of cellulose can promote the formation of intra- and intermolecular hydrogen bonds of cellulose. This special structure imparts the cellulose high hydrophilicity, which leads the cement hydration C-S-H gel to adhere to the surface of cellulosic fibers (CFs) and induce its growth. The cavity of CFs has good water absorption and can be used as an internal curing fiber for the continuous hydration of cement-based composites. But CFs in the Portland cement matrix tend to deteriorate under strong alkali conditions. This paper presents a review of the research on the durability of CFCCs. The methods and paths to improve the durability of CFCCs are summarized and analyzed from the perspectives of the internal curing of CFs, the deterioration of the performance of CFs in the matrix, and the use of many types of supplementary cementitious materials. Finally, the development and engineering application of CFCCs have been prospected.

1. Introduction

In the last few years, more and more different types of fibers have been used to reinforce cement-based composites. As is known, these fibers include steel, organic synthetic, carbon, and glass fibers. The commonly used fibers for reinforcements exhibit a set of advantages, such as long application time, relatively mature technology, and interesting physical and mechanical properties. However, compared to CFs, the commonly used fibers have their own limitations [1]. As far as we know, the density of steel fibers is relatively large, which cannot meet the requirements of reducing the weight of composites. At the same time, the size of the steel fiber is too large, there is an interface transition zone in the cement matrix, and it also lacks inducing the growth of the hydrate gel in the cement matrix interface transition zone. The organic synthetic fiber has poor compatibility with the cement matrix and is not easy to disperse in the cement matrix. It pollutes the environment greatly, which is contrary

to the concept of green cement-based composites [2–4]. The carbon fiber is easy to agglomerate in the cement matrix, and the cost is relatively high [5–8]. The glass fiber is brittle and has poor wear resistance [9, 10].

Plant fibers such as crop straws are composed of cellulose, hemicellulose, lignin, and other substances, and cellulose is the main component. CFs are widely found in nature. They are inexpensive, have low density, are renewable, are biodegradable, are rich in sources, and have good reinforcement effects on cement-based composites [11–13]. The geometric characteristics, mechanical properties, volume mixing ratio of CFs, and bonding property of the fiber and matrix interface are important factors that affect the strength and toughness of the matrix. Compared with traditional fibers, CFs have a larger specific surface area, aspect ratio, excellent toughness, and bonding ability. It disperses evenly in the cement matrix, has good compatibility, and has a filling and bridging effect on the cement matrix [14]. At the same time, the addition of CFs also greatly reduces the density of cement-based

FIGURE 1: SEM images of CFs. (a) Sisal fibers [39]. (b) Jute fibers [13]. (c, d) Combination of the macroscale fiber and cement matrix [37].

composites, improves their flexural strength [15–17], inhibits the occurrence and development of microcracks within the matrix [18, 19], and enhances the impact resistance of cement-based composites [20].

Over the last few years, studies on the toughening modification of cement-based composites using CFs have focused on the macromechanical properties of composites, the microstructure of the interface between the cement matrix and the fibers, the effect of fiber cracking and toughening, and the durability [21–24]. The durability of cement-based composites includes impermeability, frost resistance, carbonization resistance, and resistance to sulfate attack, which have an important impact on the long-term service life of the structure [25]. The durability of CFCCs is the main reason which limits their engineering application [26].

This paper presents a review of the research done in the area of the durability of CFCCs during the last years (2005.01-2021.05), which used Chinese journal literature retrieval databases, including China Journal Network and CNKI Scholar, English journal literature retrieval databases, and so on. Search keywords include cellulose fiber, concrete durability, internal curing, impermeability, frost resistance, and carbonization. In the searched literature, no relevant similar reviews have been found, and works of literature that are not related to the content of the paper have also been excluded. Only a few book chapters or reviews provide a general overview of the durability of CFCCs [27, 28], summarizing the main improvements and findings from a few research papers. There are also overviews of nanocellulose fibers, which are different from the contents discussed in this paper [29, 30]. The durability of CFCCs such as impermeability, frost resistance, and carbonization resistance is reviewed, and the impact of CFs on the durability of composites and the internal curing of CFs is discussed in this paper. From the perspectives of the performance, the degradation of CFs in the matrix, and the application of multiple types of auxiliary gel materials, a summary analysis is made to improve the durability of CFCCs, and the solutions are proposed.

2. Performance of CFs

As one of the most abundant renewable resources, CFs have the advantage of being low cost and environmentally friendly [31]. In 1932, scientists studied the chemical structure of cellulose [32]. Cellulose is a straight-chain polymer formed by linking countless D-glucopyranose anhydrides(1-5) with

$\beta(1-4)$ glycosides. The structure of cellulose is highly regular and unbranched, and the chemical formula is $(C_6H_{10}O_5)_n$. There are a large number of hydroxyl groups on the molecular chain of cellulose. The presence of this polar group promotes the formation of intra- and intermolecular hydrogen bonds of cellulose [33, 34]. CFs can be made from renewable resources such as straw, they have a porous cell structure, and their specific surface area is better than other fibers [35]. As described above, there are many pores in the cross-section of the fibers, as shown in Figures 1(a) and 1(b). The properties of CFs mainly depend on the type of cellulose. The cellulose content of different types of plants varied greatly, among which the cellulose content of cotton is the highest. Table 1 shows the performance index data of commonly used CFs and other commonly used fibers [36]. The physical and mechanical properties of CFs can be seen in Table 1. The mechanical properties of CFs such as tensile strength and Young's modulus are weaker than those of the commonly used fibers such as carbon fiber and aramid fiber. Flax, jute, ramie, sisal, and hemp fibers have better mechanical properties as similar to glass fiber in tensile strength and Young's modulus, so they can be directly used as CFCCs.

The performance of CFCCs includes working performance, mechanical properties, and durability. The effect of CFs on the working performance and mechanical properties of composites is similar to that of traditional fibers, but the impact on durability is very different. As mentioned above, the existence of a large number of hydroxyl groups on the molecular chain of cellulose promotes the formation of intra- and intermolecular hydrogen bonds of cellulose. This special structure renders the cellulose extremely hydrophilic, makes the CFs compatible with the cement matrix, and has a good cohesive force. C-S-H gel, the main hydration product of the cement, grows on the surface of CFs, as shown in Figures 1(c) and 1(d). Wu et al. [37] analyzed the SEM (Figures 1(c) and 1(d)) and found that the cement hydration around CFs was more complete. The reason is that the CFs can induce the orderly and directional growth of hydration products in the initial stage of cement hydration. After the cement hardens, when the load exceeds its cracking load, the fibers can share the load, which greatly increases the load of the cement matrix, avoiding or delaying the growth of microcracks. In Figure 1(d), the CF surface has peeled off and separated countless microfibrils with a diameter less than $1 \mu m$. Some of the microfibril ends are embedded in the cement hydration product. The formation of the

TABLE 1: Performance of CFs and commonly used fibers.

Fibers	Density (g/cm ³)	Elongation (%)	Tensile strength (MPa)	Young's modulus (GPa)
Cotton fiber	1.5-1.6	7.0-8.0	287-597	5.5-12.6
Jute fiber	1.3	1.5-1.8	393-773	26.5
Flax fiber	1.5	2.7-3.2	345-1035	27.6
Ramie fiber	1.5	3.6-3.8	400-938	61.4-128.0
Sisal fiber	1.5	2.0-2.5	511-635	9.4-22.0
Coir fiber	1.2	30.0	175	4.0-6.0
Cork fiber	1.5	—	1000	40.0
E-glass fiber	2.5	2.5	2000-3500	70.0
S-glass fiber	2.5	2.8	4570	86.0
Aramid fiber	1.4	3.3-3.7	3000-3150	63.0-67.0
Carbon fiber	1.4	1.4-1.8	4000	230.0-240.0

TABLE 2: Performance of CFs and commonly used fibers.

CFs	Length (nm)	Diameter (nm)	Tensile strength (MPa)	Young's modulus (GPa)	Ref.
Macroscale CFs (cotton)	2,000,000-3,000,000	11,540	287-597	5.5-12.6	[36]
Microcrystalline CFs	100-3,000	1,440	7,500	100.0-140.0	[37, 40, 41]
Nanocrystalline CFs	50-500	1-100			

microstructure of the cement paste will be accompanied by complex chemical reactions and physical changes. When the shrinkage of the matrix is restricted, the cement-based material will crack. The degree of cracking depends on the tensile strength and shrinkage stress of the cement paste. These microfibers play a bridging and filling role in the composites. The moisture absorbed in the CFs supplements the lack of moisture in the cement hydration process and can induce the hydration of the cement surface, and the hydration product can fill the microcracks so as to achieve the composites' effect of enhancing impermeability.

However, the lignin and hemicellulose in CFs are easily dissolved in the alkaline solution of the cement matrix, and the strong alkali material enters the fiber cavity to cause the mineralization and degradation of the fiber structure, thereby affecting the durability. The CFs applied to cement-based composites can be classified by the function of their size. CFs can be found as macroscale, microcrystalline, and nanocrystalline CFs. Macroscale CFs include strands (long fibers of lengths around 20 to 100 cm), staple fibers (short fibers with lengths between 1 and 20 cm), and pulp (very short fibers with lengths between 1 and 10 mm), which can be processed by chemical methods to form micron-scale microcrystalline CFs and nanosized nanocrystalline CFs [37, 38].

In general terms, micron-sized microcrystalline CFs and nanosized nanocrystalline CFs can be produced in chemical or enzymatic ways. The loosely arranged amorphous regions in the cellulose are destroyed, and the crystalline regions are retained to obtain micron-sized microcrystalline CFs and nanosized nanocrystalline CFs with higher crystallinity. The preparation methods of nanocrystalline CFs mainly include the physical mechanical method, acid hydrolysis method, and biological method. Nanocrystalline CFs have a rigid rod-like structure, and their properties are shown in Table 2.

3. Research Status of Durability of CFCCs

Anselme Payen, a French scientist, extracted a compound from wood in 1838 and named it cellulose. The majority of the fabrication methods for cement composites reinforced with CFs in the pulp form are based on the Hatschek process, patented by L. Hatschek in 1900 [27]. After a century, the research and application of CFCCs have become increasingly widespread. The research on CFs is from mechanical property to durability, from the bonding phenomenon between the CFs and the matrix to the bonding mechanism, and from macroscale application to microscale application. Only a few studies have been focused on the bond adhesion of CFs with cement matrices. For instance, some studies analyzed the effect of CF shape and curing age on the bond strength of CFCCs using pull-out tests [28, 42], and the other studies analyzed the fiber matrix bond adhesion indirectly [14-16].

At this stage, research studies on the durability of CFCCs include frost resistance, carbonization resistance, water penetration resistance, chloride ion penetration resistance, gas penetration resistance, sulfate erosion resistance, early anticracking performance, compressive creep performance, and compressive fatigue deformation performance. In general terms, the durability of the composites depends on the resistance to chloride ion permeability, frost resistance, and carbonization resistance. As can be seen in Table 3, it is a summary of related studies on the durability of CFCCs.

Table 3 shows that CFCCs are rich in varieties, and their durability involved a wide range, which are closely related to the service life of the composites. The test methods for resistance of CFCCs to freezing and thawing usually include slow freezing and thawing, rapid freezing and thawing, and single-side freezing and thawing. The test methods for resistance of CFCCs to chloride penetration adopt the rapid chloride

TABLE 3: Summary of studies on durability of CFCCs.

Durability	CFs	CFs (wt.%)	Fiber form	Cementitious	Test methods	Ref.
Antichloride ion penetration	UF500 CF	0.45~1.5	Randomly dispersed	P-O42.5	RCM method; water seepage height	[43–45]
	Bokai super fiber	0.6~1.2		P-O52.5	Water seepage pressure	[46]
Antifreezing	Flake fiber	0.9	Randomly dispersed	P-O42.5	Rapid freezing	[43, 47]
	UF500 CF	0.1~0.2				[48]
Carbonization	Cork kraft pulp	8.0	Aligned	P-II42.5R	Accelerate carbonization	[49]
	Rape straw fiber	0.5~2.00	Randomly dispersed	SAC		[50, 51]
	Coir fiber	2.0		Slag cement	12 years	[52]
	Sisal fiber	1.5~8.0	Randomly dispersed	P-O42.5R	CBI method	[53, 54]
Antidry and wet cycle	Cork CF	4.0		P-O42.5	25 dry/wet cycles	[55]
	Sisal fiber	0.6	Aligned	Calcined clay	100 dry/wet cycles	[56]
Antisulfate attack	UF500 CF	0.9	Randomly dispersed	P-II42.5R	Sulfate-wet and dry cycle coupling	[51]
Anticrack	Sisal fiber	2.0	Aligned	MK/PC	Instron 5948 test system	[57]

migration (RCM) coefficient test and coulomb electric flux test. The procedures for preparing CFCCs reported in the literature can be divided into two main groups depending on the fiber form: fibers randomly dispersed in the cement matrix [58–60] and aligned fibers or fibrous structures [43, 46, 50, 53]. In view of the fibers randomly dispersed in the matrix, it has certain limitations in the mechanical properties of reinforced composites, and aligned fibers or fibrous structures are also used to strengthen the cement matrix [49, 56, 57, 61].

Cement-based composite is a kind of porous material, which provides a channel for harmful external impurities to penetrate into the matrix. Adding CFs to the cement matrix can reduce the generation of early microcracks, inhibit the development of microcracks during the service period of the matrix, and improve the durability of CFCCs. After absorbing water, the CFs are evenly dispersed into the cement matrix, forming a plurality of microwater flow channels inside the matrix. These pores can continue to provide water for the later cement hydration, make it fully hydrated, ensure the mechanical properties of the cement matrix, prevent the cement matrix from cracking, and also improve the cement matrix's antipermeability, antifreezing, and anticarbonization capabilities. The uniform distribution of the fiber network enhances the adhesion between the matrix components, the matrix structure has good integrity, and the impact resistance is also significantly improved [62].

3.1. Impermeability of CFCCs. Impermeability is an important factor that affects the durability of CFCCs. CFs are evenly distributed in the cement matrix, which can reduce the segregation effect in the initial stage of the cement hydration, inhibit the formation of shrinkage cracks in the cement matrix, reduce the porosity of the matrix, improve the compactness of the matrix, and effectively prevent harmful substances from penetrating into the matrix. It is usually determined by the rapid chloride ion migration coefficient test or the water penetration height test [44]. The chloride ion diffusion coefficient and the water penetration height are reduced after the CFs have been mixed, and the imperme-

ability of the CFs is better than that of the polypropylene fibers under the same dosage. The test results showed that when the CF volume fraction was 0.9%, the effect of improving the impermeability of the matrix was the best [45]. CFs have little effect on the compressive strength of the matrix but significantly improve the splitting tensile strength, axial tensile strength, and ultimate tensile value and effectively block the penetration of chloride ions. At the same time, the cracking time and the width of the matrix crack have been improved [63, 64]. It has been found through experiments that the bubble content in the CF cement matrix decreases by 40%, and the bubble spacing becomes smaller [65].

CFs are effectively bonded to the cement matrix and distributed in random directions, forming a uniform support system, optimizing the pore structure of the cement matrix, and blocking the internal communication channels of the matrix. Due to the toughening and cracking resistance of CFs, the number of initial cracks can be significantly reduced, the long-term cracks of the matrix can be effectively inhibited, and the possibility of forming through cracks in the matrix can be reduced. In this way, the microcrack pattern of the cement-based composite is further refined, and its impermeability is significantly improved.

3.2. Effect of Freeze-Thaw Cycles on the Durability of CFCCs.

Due to the effect of negative temperature, the water in the pores of CFCCs in a water-saturated state freezes and produces a volume change to form tensile stress. Under the action of the freeze-thaw cycle, the cement matrix damage gradually accumulates and expands, which will eventually lead to the destruction of the cement matrix. CFs can significantly improve the frost resistance of CFCCs. On the one hand, the addition of CFs reduces the water penetration in the cement matrix. On the other hand, the CFs can absorb parts of the unfrozen free water and reduce the hydrostatic pressure in the cement matrix. A large number of comparative tests have shown that polypropylene fibers and CFs have improved frost resistance to cement-based composites [47, 66, 67], and CFs are better than polypropylene fibers.

FIGURE 2: (a) The relative dynamic elastic modulus of basalt fibers and (b) CF with different fiber volume fraction changes with the number of freeze-thaw cycles [48].

When the volume fraction of CFs is 0.9%, the antifreezing effect of CFCCs is the best. Under the action of the freeze-thaw cycle, the CFs have the effect of binding the surface slurry of the matrix, and its appearance damage is improved to a certain extent.

The addition of CFs slows down the rate of decrease of relative dynamic elastic modulus, increases the number of freeze-thaw cycles that the test piece can withstand, and improves the frost resistance of the CFCCs; it can be seen from Figure 2 [48]. As the number of freeze-thaw cycles increases, the relative dynamic elastic modulus of the specimens decreases. At the beginning of the freeze-thaw cycle (within 100 freeze-thaw cycles), for basalt fibers and CFs, when the fiber volume fraction is 1.0, 1.5, or 2.0%, the relative dynamic elastic modulus decreases slowly, and the effect of fiber content is not much different. In the late freeze-thaw period (the number of freeze-thaw cycles is greater than 100), the decline rate increases, indicating that the internal damage of the concrete gradually increases after the freeze-thaw cycle. The relative dynamic elastic modulus of plain concrete decreases faster than that of fiber concrete. When the freeze-thaw cycle reaches 200 times, the freeze-thaw damage rate is far less than 60%. However, the relative dynamic elastic modulus of fiber-reinforced concrete with a fiber volume fraction of 2% decreases more gently than that of concrete with fiber volume fractions of 1.0% and 1.5%.

Since the elastic modulus and tensile properties of basalt fibers are higher than those of CFs, the basalt fibers can more effectively improve the tensile strength of concrete, inhibit the expansion of internal cracks in concrete, reduce the entry of water into the matrix, and delay the frost heave damage of the internal structure. Therefore, when the number of freeze-thaw cycles is high, basalt fibers improve the freeze resistance of concrete more significantly than CFs. In general, the effect of improving the frost resistance is not significant when the fiber volume fraction is increased from 1.5% to 2.0%. Therefore, it is more economical and reasonable to choose the fiber volume fraction of 1.5%.

3.3. Effect of Carbonization on the Durability of CFCCs.

When CFCCs are applied as the protective layer of steel bars, the impact of carbonization on the steel bars must be paid attention to. The high alkalinity inside the matrix passivates the surface of the steel bar, and the passivated film can prevent the steel bar from corroding by the external environment. The hydration product of cement-based composites has stable performance in an alkaline environment and can maintain good cementing ability. Carbonization is the process of neutralizing the cement matrix, which can reduce the alkalinity of CFCCs and can induce the corrosion of the stressed steel bars and the destruction of the structure [68, 69]. The essence of carbonization is the diffusion process of carbon dioxide gas from the surface to the inside of the matrix. Both compactness of the matrix structure and internal defects affect the diffusion rate. The hydrophilicity and unique hollow structure of CFs optimize the pore structure of cement-based composites, which reduce internal defects and enhance carbonization resistance. In the case of loading, the generated microcracks become channels for the diffusion of carbon dioxide gas, which reduce its anticarbonization performance. After adding CFs to the matrix, the carbonization of cement-based composites is still a diffusion-dominated process [70–72].

Analysis of the pore size distribution data obtained based on the mercury intrusion experiment method shows that the fiber has a significant influence on the microstructure of cement-based composites [73]. After the fiber volume fraction reaches 0.6%, it can be observed that the unimodal width on the pore distribution curve of cement-based materials increases, and the number of large-scale pores (radius greater than or equal to 200 nanometers) increases, which confirms that the micromatrix structure is coarsened due to the introduction of fibers. The addition of fibers can change the working performance of the fresh cement paste, thereby producing an air-entraining effect [74]. At the 0.6% fiber volume fraction, the effect of this phenomenon exceeds the internal curing effect, which causes the degradation of the

FIGURE 3: (a) Influence of fiber content on fracture toughness and (b) influence of fiber content on composite deformation [78].

TABLE 4: Bulk density of CFCCs at 28 days (g/cm³).

CFs	Fiber content (wt.%)				
	0	4	8	12	16
RFRCC	2.01 ± 0.04	1.70 ± 0.01	1.49 ± 0.03	1.38 ± 0.04	1.26 ± 0.01
BFRCC	2.01 ± 0.04	1.71 ± 0.01	1.50 ± 0.04	1.36 ± 0.04	1.30 ± 0.05

overall microstructure of cement-based composites. Compared with the 0.6% fiber volume fraction, at the 0.3% fiber volume fraction, the pore size distribution before and after carbonization is less different, indicating that the 0.3% fiber volume fraction has a limited effect on the microstructure of cement-based composites.

4. Constitutive Relationship between CFs and Composite Durability

According to the fiber spacing theory proposed by the concept of fiber crack resistance, the use of densely spaced fibers as a crack barrier can reduce the stress intensity factor of the microcrack tip inside the matrix and inhibit the propagation of microcracks in the matrix, thereby improving the initial cracking strength of the composites. It is known from the previous studies that there is a certain limit on the amount of CF, which has a significant impact on the durability of composites due to the internal curing of the cement matrix and the long-term alkaline environment.

In fact, CFs increase the air content of the concrete and relieve the hydrostatic pressure and osmotic pressure during low-temperature cycles. Secondly, dense microfibers improve the internal quality of the concrete, reduce internal defects, and improve concrete's tensile properties such as ultimate tensile strain and fracture energy. In addition, due to the small diameter of CFs and the large number of fibers per unit weight, the fiber spacing is small, which increases the energy

loss during the concrete damage process and effectively inhibits the cracking of the concrete.

4.1. Internal Curing Fiber of Cement-Based Composites. Different from other types of fibers, CFs have a unique hollow lumen structure and good water absorption. It can be used as the internal curing fiber of cement-based composites, as shown in Figures 1(a) and 1(b). In the absence of an external water supply for maintenance, it can play its role in the internal maintenance of the cement matrix, improve the water loss of the cement matrix under natural conditions, and promote continuous hydration for a long time. Therefore, its later strength increases greatly [75, 76]. CFs can also harmonize the workability of composites and improve construction performance. In addition, the use of the curing properties of CFs can improve the interlayer deposition and stacking process of 3D printed cement-based composites, reduce interlayer voids and longitudinal defects, and enhance durability [77].

CFs can effectively reduce the shrinkage of the cement matrix and significantly improve the flexural strength and fracture toughness of composites. The 28-day fracture toughness and 100-day fracture toughness of the composite vary with the fiber content, as shown in Figure 3(a). The physical parameter of CFCCs varies with fiber content. As the amount of CFs increases, the density of the composites decreases. Table 4 shows the density of CFCCs [78]. When the fiber content is 16% by mass, the 28-day fracture toughness is increased by 37 times. Figure 3(b) shows the relationship

FIGURE 4: (a) Microstructure [72]. (b) Voids around the CFs. (c) Micro-nano-level microcrystalline cellulose [35].

between the deflection and the fiber content of two CFCCs of rice straw (RFRCC) and bamboo (BFRCC) [78]. It can be seen that as the CF content increased, the deflection of the test piece also increased, which further shows that the fiber improved the deformability and toughness of the composite, thereby enhancing the durability of the composites. As shown in Figure 3, composites reinforced with CFs experience a significant reduction in fracture toughness along with the prolongation of time. This means that the CF-reinforced composites will become stiffer and more brittle with time. Melo Filho and his coworkers [39] suggested that the weakening of energy absorbability of the CF was probably due to the deposition of calcium hydroxide crystals on the CF surface.

4.2. Deterioration of CFs in the Cement Matrix. The structural characteristics of CFs are the root cause of their deterioration in the environment of a high alkaline cement matrix. Studies have shown that the fiber is in an alkaline environment for a long time, and the lignin and hemicellulose in fibers are easy to dissolve in the alkaline solution of the cement-based composite, resulting in partial fiber breakage and tensile strength weakening. On the other hand, the strong alkali substance of the cement matrix enters into the fiber cavity to cause the mineralization of the fiber structure, which can reduce the mechanical properties of the fiber. At the same time, the extremely strong hydrophilicity of CFs causes its volume to change, which affects the overall structure durability [49, 57]. The sisal fiber and coconut husk fiber were immersed in a calcium hydroxide-saturated solution, the strength test was conducted for 28 days, and it was found that the tensile strength decreased by about 50% [52]. When CFs are immersed in water, saturated lime water, and sodium hydroxide solution, the lignin, cellulose, and hemicellulose contents of the fibers are all reduced [37, 58]. The application of these deteriorated fibers in cement-based composites will inevitably cause the mechanical properties of cement-based composites to decrease.

4.3. Ways to Improve the Durability of CFCCs. At present, there are generally two methods to improve the durability

of CFCCs based on the internal curing and prone to deterioration characteristics of CFs. One method is to modify the cement matrix to consume the calcium hydroxide content of the alkaline component produced during cement hydration. Another method is to modify the fibers to improve the stability of the fibers in the cement matrix by physical or chemical methods.

4.3.1. Modification of the Cement Matrix. The reinforced concrete structure should be prevented from carbonization, and as for cement-based composites reinforced with single CFs, carbonization needs to be accelerated in order to enhance their durability. The purpose of carbonization is to make the cement hydration product calcium hydroxide react with carbon dioxide to form calcium carbonate. Pizzol et al. [79] have done the strengthened and accelerated carbonization test of the composite with sisal and kraft pulp, which increased the load-bearing capacity of the composite by 25% and the toughness by 80%, and reduced the fiber degradation in the cement medium. Carbonization reduced the porosity, water absorption, and nitrogen permeability of the composite, increased the matrix interface density, and made the fiber and cement matrix bond tighter, as shown in Figure 4(a). Carbonization improved the compressive strength and the durability and weather resistance of the composites, and their service life was extended [70, 71, 80]. Due to the chemical stability of the carbonized product and its reduced capillary porosity, the CFCCs have better flexural strength and can improve the adhesion between the cement-based matrix and the CFs.

Studies have shown that the optimal water content of the carbonized matrix is 40% to 60% [72], and carbonization significantly improves the durability of the matrix against dry and wet freezing and thawing. Both carbonization and the addition of mineral admixtures can reduce the calcium hydroxide content in the cement matrix. The cement-based composite is mixed with mineral admixtures such as silica fume, metakaolin, blast furnace slag, and fly ash, which can undergo secondary hydration reaction with calcium hydroxide in the cement to obtain hydrated calcium silicate or hydrated calcium aluminate [81]. Replacing part of cement

TABLE 5: Use of supplementary cementitious materials of CFCCs.

Cementitious materials	Weight of cement (%)	CFs	Extent of improvement	Ref.
MK	50 MK	Sisal	Significant reduction of the calcium hydroxide formation; no signs of fiber degradation	[38, 64, 83]
MK and SF	15 SF or 15 MK	Sisal	Improved the mechanical properties and the durability	[81]
SL, SF, and MK	70 SL/10 MK or 70 SL/10 SF	Kraft pulp	Effective in preventing degradation	[82]
RHA, MK, and NC	30 RHA, MK, and NC	Sisal	The durability of composites was improved owing to the mitigation of fiber degradation	[53]
SF and SL	10 SF and 40% SL	Cannabinus	Slowing down the strength loss and embrittlement	[84]
SF, SL, FA, MK	10% SF/70% SL, 10% MK/70% SL, and 10% MK/10% SF/70% FA	Softwood kraft pulp	Prevented composite degradation due to a reduction in the calcium hydroxide content and the stabilization of the alkali content	[85]
SL, GY, LI	88 SL/10 GY/2 LI (0 cement)	Coir and sisal	Do not appear to have a significant effect on the prevention of ductility dropping	[86]
SF, NRL	13.55 SF/14.55 CF/1.40 NRL	Cellulose	Improved material durability	[20]

TABLE 6: Modification of CFs and the extent of improvement.

Type	Method	CFs	Extent of improvement	Ref.
Physical modification	Polyelectrolyte adsorption	Blue eucalyptus paper	Antibacterial effect	[91]
	Nonelectrolyte adsorption	Cellulose nanocrystals	Changed the surface structure and properties of cellulose	[92]
	Small molecule modification	Pulp	Promoted the preparation of nanocellulose	[93]
Chemical modification	Graft modification	Softwood cellulose fiber	Formed a strong nanocomposite	[94]
	Cross-linking modification	Cellulose nanocrystals	Improved thermal stability and water resistance, decreased swelling degree	[95]
Biological modification	<i>In situ</i> modification	6-Carboxyfluorescein-modified glucose	With nonnatural characteristic fluorescent function	[96]
	<i>Ex situ</i> modification	Proanthocyanidins as cross-linking agent	Novel bacterial cellulose/gelatin composite	[97, 98]

with mineral admixtures significantly reduces the content of calcium hydroxide, avoids the deterioration of fiber performance, and ensures the strength and toughness of cement-based composites [82]. More studies revealed that it can produce cementitious materials without calcium hydroxide by using calcined metakaolin and calcined waste crushed clay bricks instead of ordinary Portland cement [53].

There are many types of supplementary cementitious materials used, and the extent of improvement varies. As shown in Table 5, the abbreviations of each component are as follows: silica fume (SF), blast furnace slag (SL), fly ash (FA), metakaolin (MK), rice husk ash (RHA), natural rubber latex (NRL), nanoclay (NC), gypsum (GY), and lime (LI).

4.3.2. Modification of CFs. Improving the water resistance of CFs and the adhesion between the matrix and the fiber interface is a necessary method to develop composites with good mechanical and environmental properties. However, the various types of CFs, geographical and climatic conditions, and growth cycles make the performance of CFs different. Some CFs have poor chemical resistance and low strength. These

fibers can be modified to improve their internal and external structures and mechanical performance. The modification of CFs mainly includes physical modification, chemical modification, and biological modification, among which chemical modification is the most common [87–90], as shown in Table 6.

In the modification method of cellulose, the physical method is simple, convenient, and easy to operate, but the performance of the modified product is unstable, and the modifier is easy to fall off from the cellulose, resulting in product performance reduction. The chemical method is a better modification method [91, 92]. Compared with small molecule modification, graft polymerization has obvious advantages. It imparts other properties to cellulose without changing the properties of cellulose, and the modification effect is very stable. However, there are also disadvantages such as difficulty in operation and difficulty in controlling the reaction [93–95]. The biological modification to cellulose should be modified *in situ* or *ex situ* according to the actual situation [96–98].

The fibers are chemically modified to remove hemicellulose, lignin, pectin, and other substances on its surface so that

the structure of CFs becomes fibrillation and, which have a relatively rough appearance. The cement matrix interface forms a mechanical interlocking morphology [99–101].

When eucalyptus fibers are modified with 3-mercaptopropyltrimethoxysilane [102], it is found that the fiber reduces the water retention and meanwhile improves the dimensional stability of the composite. Through the dry-wet cycle treatment of abaca, agave, and sisal fibers [103], the cross-section of the fiber is reduced, Young's modulus is increased, and the tensile strength and tensile strain are reduced. At the same time, the cavity becomes thinner. The modified fibers increase the interfacial shear strength of the cement-based composite and also improve the durability. By adding 5% styrene-acrylic copolymer for treatment, after 200 dry and wet cycles, the water absorption ratio of the test piece is reduced by 50%, the elastic modulus value is reduced by 40%, and the shrinkage rate is reduced by 15%, which improves the stiffness and dimensional stability of the specimen [104]. After the modification treatment, the interface between the fibers and the cement matrix forms a dense and cohesive transition zone, which makes the fiber adhere to the cement surface to prevent the fiber from mineralizing.

4.3.3. Multitype CFs. Macroscale CFs are of large diameter and cavity, so they will absorb water and swell in the initial stage of mixing with the cement matrix. In the later stage of cement hydration, the fiber moisture gradually loses, the fiber shrinks and collapses in the matrix interface, and some voids are left at the fiber and cement matrix interface, which affects the performance of the composite, as shown in Figure 4(b). In order to improve this situation, the acid hydrolysis method can be used to prepare micron-sized microcrystalline CFs and nanosized nanocrystalline CFs [105, 106].

Microcrystalline CF is super absorbent, which can supplement the lack of moisture in the cement matrix at the later stage of hydration so that the cement matrix can be fully hydrated. A large amount of cement hydration gel can be induced around the microcrystalline CFs in the cement matrix to fill the microcracks and voids of the matrix and reduce the dry shrinkage cracks of the cement matrix at the initial stage of hydration. Nanocrystalline CF is of the same size as the cement hydration gel, which can induce the cement matrix hydration C-S-H gel to adhere to the surface of the nanocrystalline CFs, so they are connected and fused to form a uniform continuous C-S-H gel phase in the cement matrix. The cement matrix hydrate can completely embed in the nanocrystalline CFs, avoiding the negative effects of volume instability caused by macroscale CFs and microcrystalline CFs, and can further improve the durability of the composite, as shown in Figure 4(c). The shrinkage rate and mechanical properties of the material have been improved by using micro-nano-microcrystalline cellulose to toughen the concrete [107, 108]. Compared with ordinary samples, the network structure formed by the multitype fibers can transmit and share the stress generated by the plastic shrinkage of the cement matrix. The combination of the fiber and the matrix improved the crack resistance of the material and also enhanced its durability.

5. Conclusions

When microcracks appear during the service period of cement-based composites, the fibers share the load through the bridging action, which slows down the continuous development of the microcracks and increases the durability of the composites. The main conclusions are as follows.

The hydrophilicity and unique hollow structure of CFs optimize the pore structure of cement-based composites, so CFs can significantly improve the permeability, frost resistance, and carbonization resistance of CFCCs.

CFs uniformly dispersed in the cement matrix, and it can induce the orderly growth of cement hydration products at the initial stage of hydration and enhance the compactness of the cement matrix.

The internal curing characteristics of CFs on the cement matrix can enhance the durability of CFCCs. The utilization of microcrystalline CFs and nanocrystalline CFs has further improved the durability of CFCCs.

Cementitious materials with low alkali corrosion have been used to reduce the long-term performance degradation of CFs, such as magnesium silicate cement, magnesium phosphate cement, and geopolymer cement.

Fiber modification is an important measure to improve the durability of CFCCs, and in particular, chemical modification has been usually used.

Data Availability

All underlying data are provided in full within this paper.

Disclosure

The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

Conflicts of Interest

The authors declare no conflict of interest.

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